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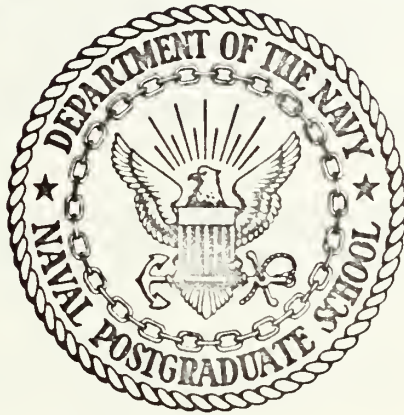
THE EXCITATION OF NITROGEN FOR THE FIRST  
NEGATIVE AND SECOND POSITIVE SYSTEMS BY  
HIGH ENERGY PROTON BOMBARDMENT

Enrique J. Vera



# NAVAL POSTGRADUATE SCHOOL

## Monterey, California



# THESIS

THE EXCITATION OF NITROGEN  
FOR THE FIRST NEGATIVE AND SECOND POSITIVE SYSTEMS  
BY HIGH ENERGY PROTON BOMBARDMENT

by

Enrique J. Vera

Thesis Advisor:

E.A. Milne

June 1972

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The Excitation of Nitrogen  
For the First Negative and Second Positive Systems  
By High Energy Proton Bombardment

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MASTER OF SCIENCE IN PHYSICS

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## ABSTRACT

Nitrogen and nitrogen-helium mixtures were excited by 1.4 MeV protons from a Van de Graaff generator. Intensity versus pressure data from 1 to 600 Torr were recorded and plotted for the first negative band, transitions  $v = 0$  to  $v' = 0$  ( $\lambda = 3914.4 \text{ \AA}$ ), and  $v = 0$  to  $v' = 1$  ( $\lambda = 4278.1 \text{ \AA}$ ) and second positive transition  $v = 0$  to  $v' = 0$  ( $\lambda = 3371.3 \text{ \AA}$ ) of molecular nitrogen. The theoretical equation from Smelley's work [1] for nitrogen alone was verified and the coefficient  $\frac{B}{A}$  from Smelley's work was used in this work. A theoretical equation was derived for intensity as a function of partial pressure of nitrogen and helium, which was shown to agree quite well with the experimental data and the coefficient of enhancement  $C = 0.251 \pm 0.001$  was obtained. As the experimental data shows, a very interesting collisional excitation process was observed, which was not observed for nitrogen-oxygen and nitrogen-carbon dioxide mixtures.





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## ACKNOWLEDGEMENT

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## I. INTRODUCTION

Studies on the excitation of nitrogen ( $N_2$ ) by proton impact have been done by several investigators, and measurements were made to determine the de-excitation reaction rate and the de-excitation cross sections for ( $N_2$ )\* second positive ( $C^3\Pi_u \rightarrow B^3\Pi_g$ ) and ( $N_2^+$ )\* first negative ( $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ ) transitions.

Since, the problem in consideration is of interest in connection with the phenomena of upper atmosphere, such as the mechanisms for production of excited states by energetic particles and the ways in which the energy is contained in excitation and transmitted through collision, a continuation of these studies has been done [1-2]. This work was a continuation of these studies, using similar experimental procedure to investigate the effect of helium (He) on the nitrogen ( $N_2$ ) gas system. In this study the pressure was varied from 1 to 600 Torr keeping the partial pressure of nitrogen fixed at 100, 50 or 10 Torr for each transition observed, adding helium (He) in increments up to the total pressure of 800 Torr. The proton energy was fixed at 1.5 MeV.

This experiment revealed that the quenching effects, observed in the preceding experiments of mixtures of other gases with nitrogen ( $N_2$ ) such as carbon dioxide ( $CO_2$ ), were not present, on the contrary an enhancement of the second positive and first negative transitions of nitrogen were observed.





## II. EXPERIMENTAL PROCEDURE

A 2.0 MeV Van de Graaff generator was used to accelerate the protons used to excite the gas contained in the collision chamber. The collision chamber, optical apparatus, and electronics schematic are shown in Figure 1. To separate the vacuum of the Van de Graaff from the gas in the collision chamber, a thin aluminum window of  $5.0 \times 10^{-4}$  inches thick was placed between them. The target gas could then be varied without affecting the vacuum in the Van de Graaff. The energy loss in passing through the aluminum foil is  $0.25 \pm 0.05$  MeV [1]. The reaction chamber, made of a pyrex "T" is attached to a faraday cup which collects the proton beam. The target gas was fed into the reaction chamber through the manifold, which is shown in Figure 2. The pressure in the chamber was measured by two Wallace and Tiernan gauges, one with a 0-50 Torr scale and the other with a 0-800 Torr scale, shown in Figure 3. The reaction chamber can be evacuated by a liquid nitrogen trapped oil diffusion pump to  $4 \times 10^{-7}$  Torr.

The proton beam current collected in the Faraday cup was measured and integrated by an Eldorado Electronics current integrator, model CI-110.

The radiation from the excited target gas was focused by a 15 cm focal length quartz lens located at an angle of 90 degrees from the axis of the proton beam, through a mechanical chopper, into the entrance slits of the Jerral Ash monochromator. The resolving power of the monochromator for 250 micron slits was



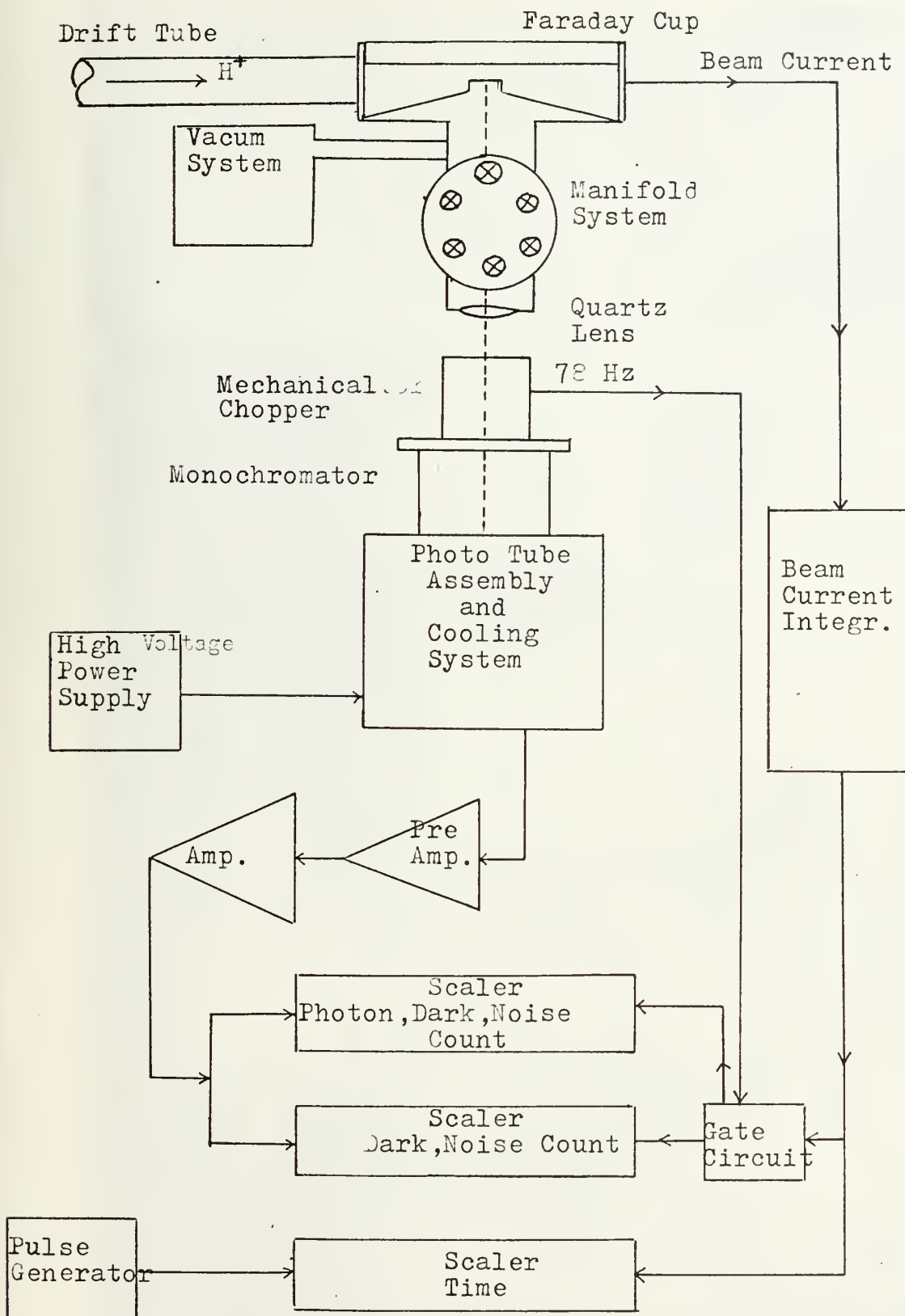


Fig. 1. Collision Chamber, Optical Apparatus, and Electronics Schematic





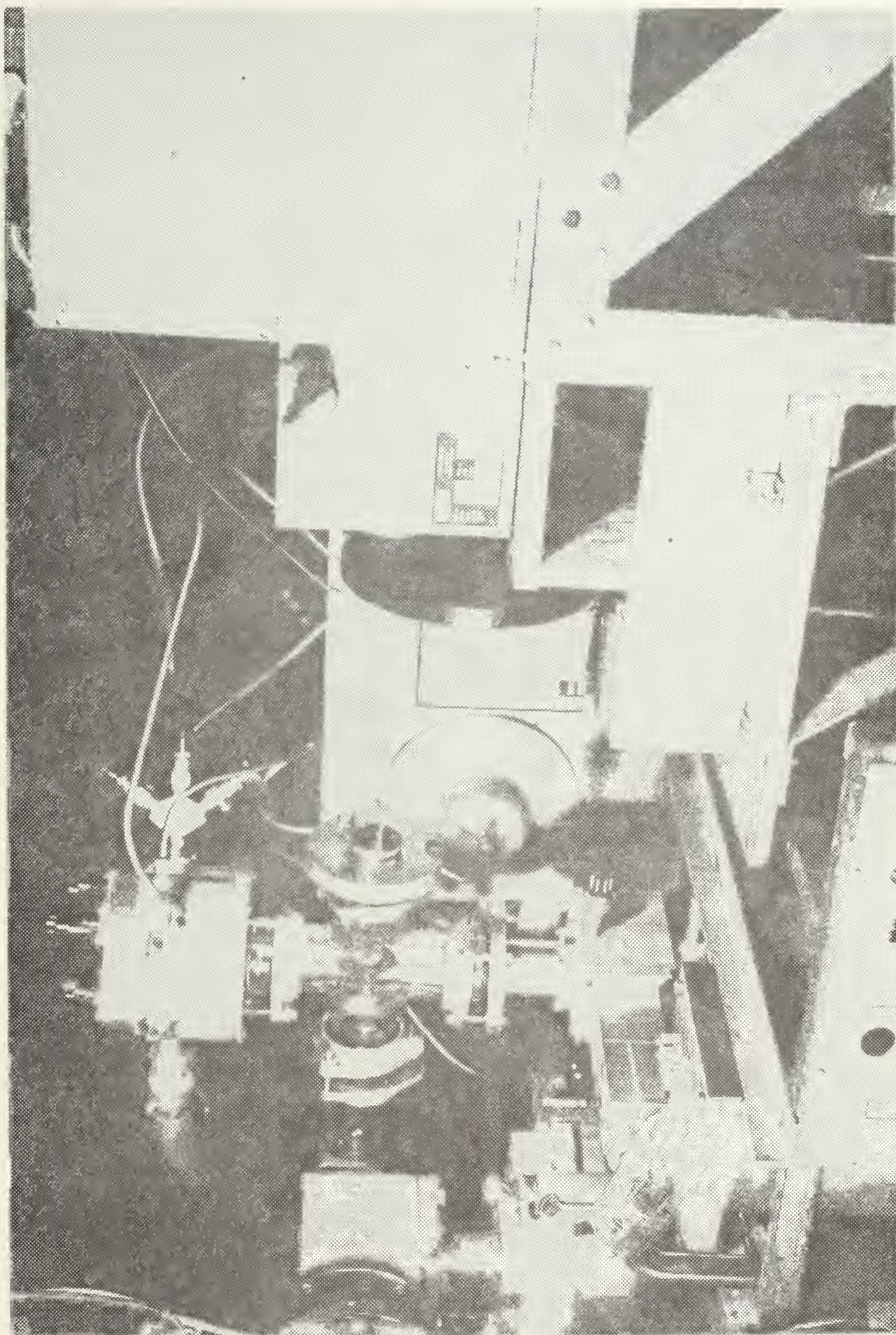


Figure 2. Collision Chamber, Control Manifold and Optical Equipment.





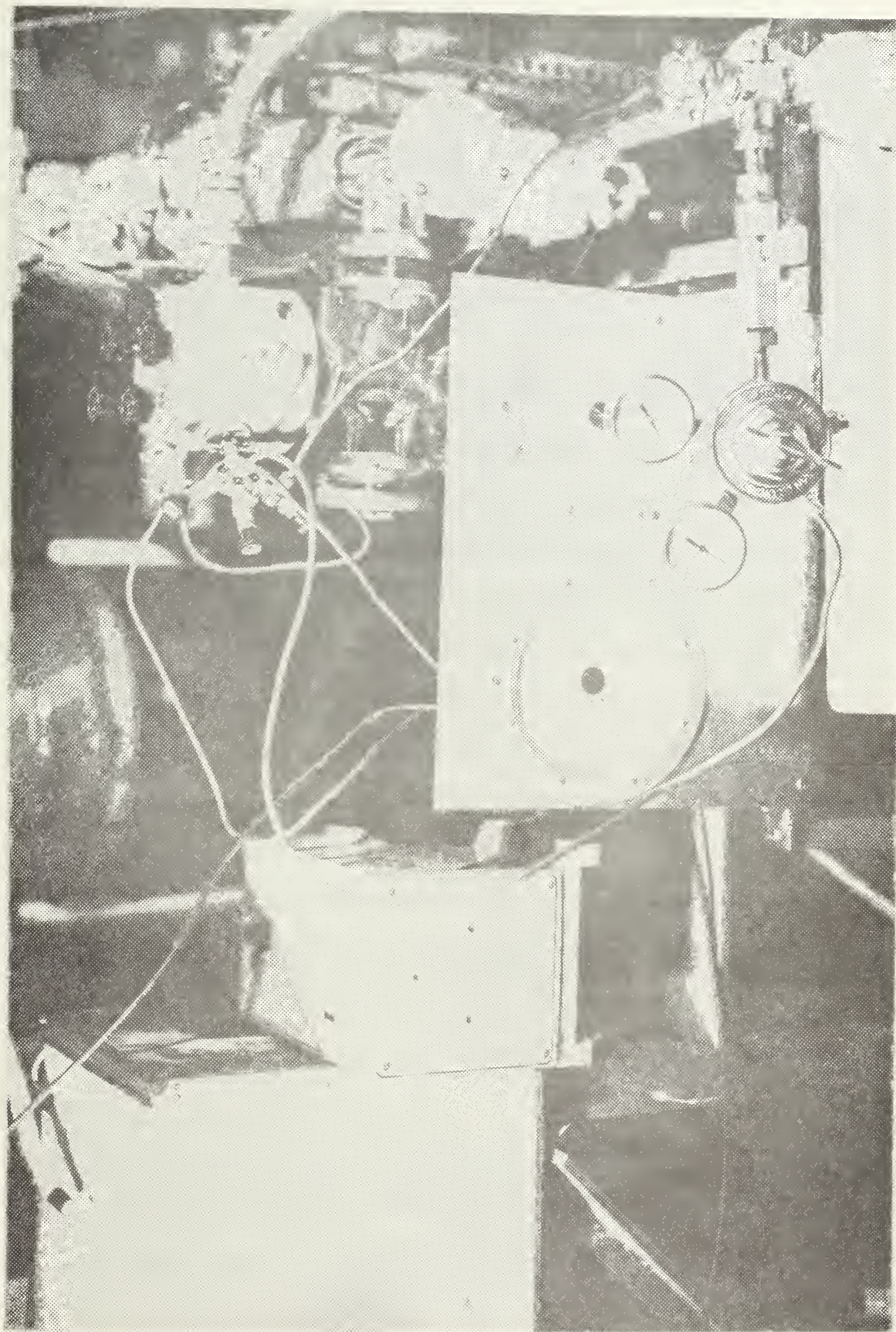


Figure 3. Pressure Gages, Monochromator and Photo Tube Assembly.





$\pm 8$  A, measured using a mercury spectrum. The light passes into a photomultiplier tube assembly where the light was converted into an electrical signal proportional to the light's intensity, see Figure 4. The electrical signal was fed into a Princeton Applied Research model HR-8, lock-in amplifier, where the signal was compared to the mechanical chopper's reference signal. The monochromator was set on the desired wavelength prior to an experimental run.

The output signal of the photomultiplier tube went to a preamplifier, amplifier, and a discriminator, and then to two scalers. The scalers were gated so that one scaler counted during the period of time that the photomultiplier tube received light, and the other scaler counted during the time that the mechanical chopper interrupted the light. Both scalers were controlled by a transistorized gate circuit which received signals from the beam current integrator and the 78 Hz reference from the chopper. Another scaler was pulsed at a rate of one pulse every 0.1 second and also gated by the current integrator. The beam current integrator was set so that when 20 microcoulombs of charge were collected from the beam, the counting and timer scalers were stopped. Thus one scaler counted photons and background, the second scaler counted background and the third scaler recorded the time.

As was described by R.F. Walters [2], the intensity ( $I$ ) is proportional to the photon count ( $NC$ ), which has been corrected for background, and inversely proportional to the total proton





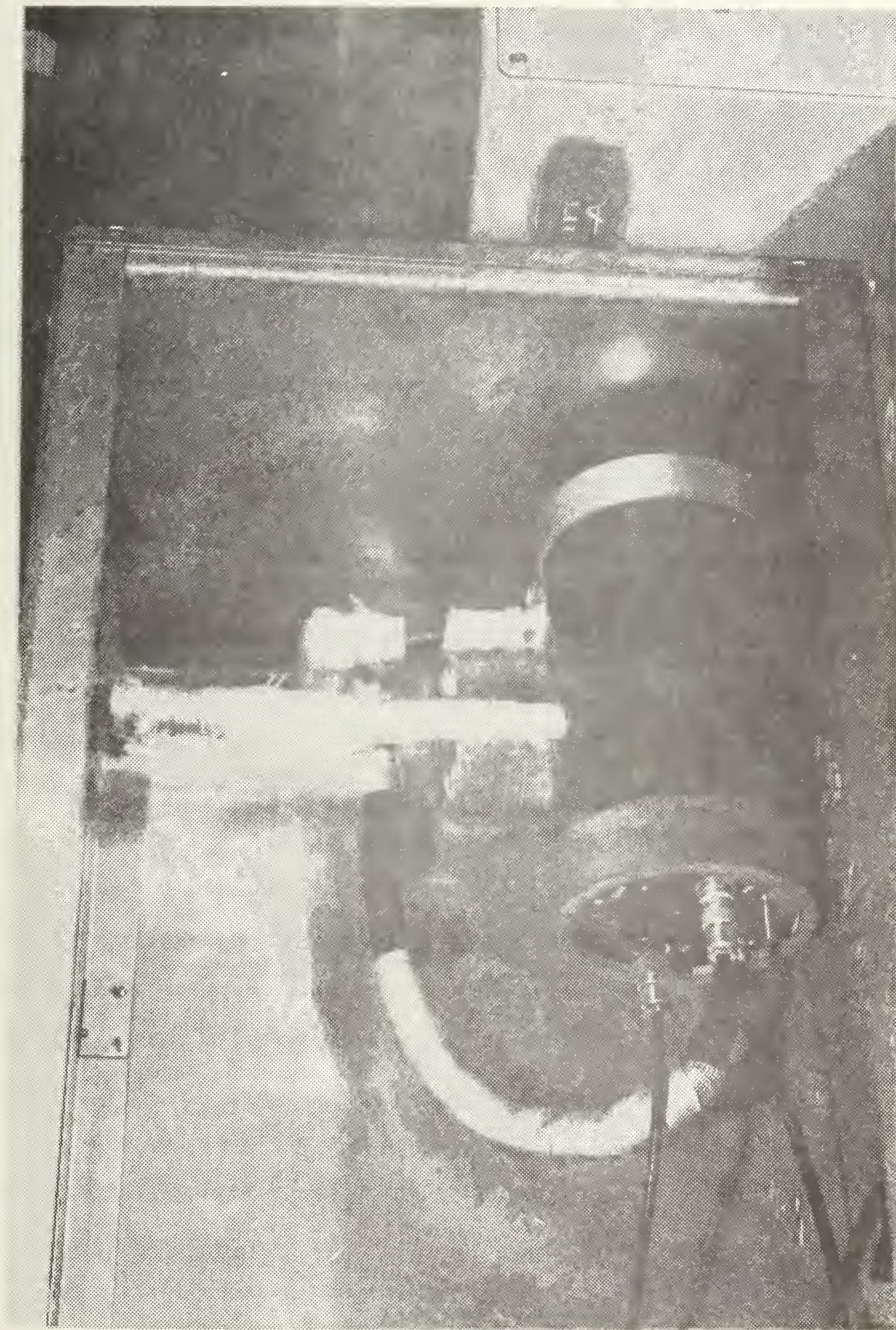


Figure 4. Photo Tube and Cooling System.





beam charge collected (QB). Then the equations are:

$$I = NC/QB \quad (1)$$

$$NC = N_{ic} - N_2 \quad (2)$$

$$N_{lc} = \frac{N_1}{[1.0 - (N_1 \times t/T)]} \quad (3)$$

where,

$N_{lc}$  = count of photons plus background corrected for dead time of system

$N_1$  = count of photons plus background

$N_2$  = background count

$T$  = counting time in seconds

$t$  = dead time of the system =  $8 \times 10^{-7}$  sec.

The value of QB was fixed at 20 microcoulombs of charge. The counting time was the time required to collect this charge.

As mentioned before, the energy of the proton beam was set at 1.5 MeV and the average target current was 1.5 microamperes.

During the observation of each run, care was taken to ensure that the gases were well mixed by waiting four minutes each time that helium (He) was added to the chamber before making a count. The temperature of the reacting molecules is assured to remain constant [2].

Smelley [1] reported excellent results in photographing the nitrogen ( $N_2$ ) spectrum and noted that  $3371.3\text{\AA}$  and  $3914\text{\AA}$  lines were the most intense.

The control console and the measuring devices are shown in Figure 5.



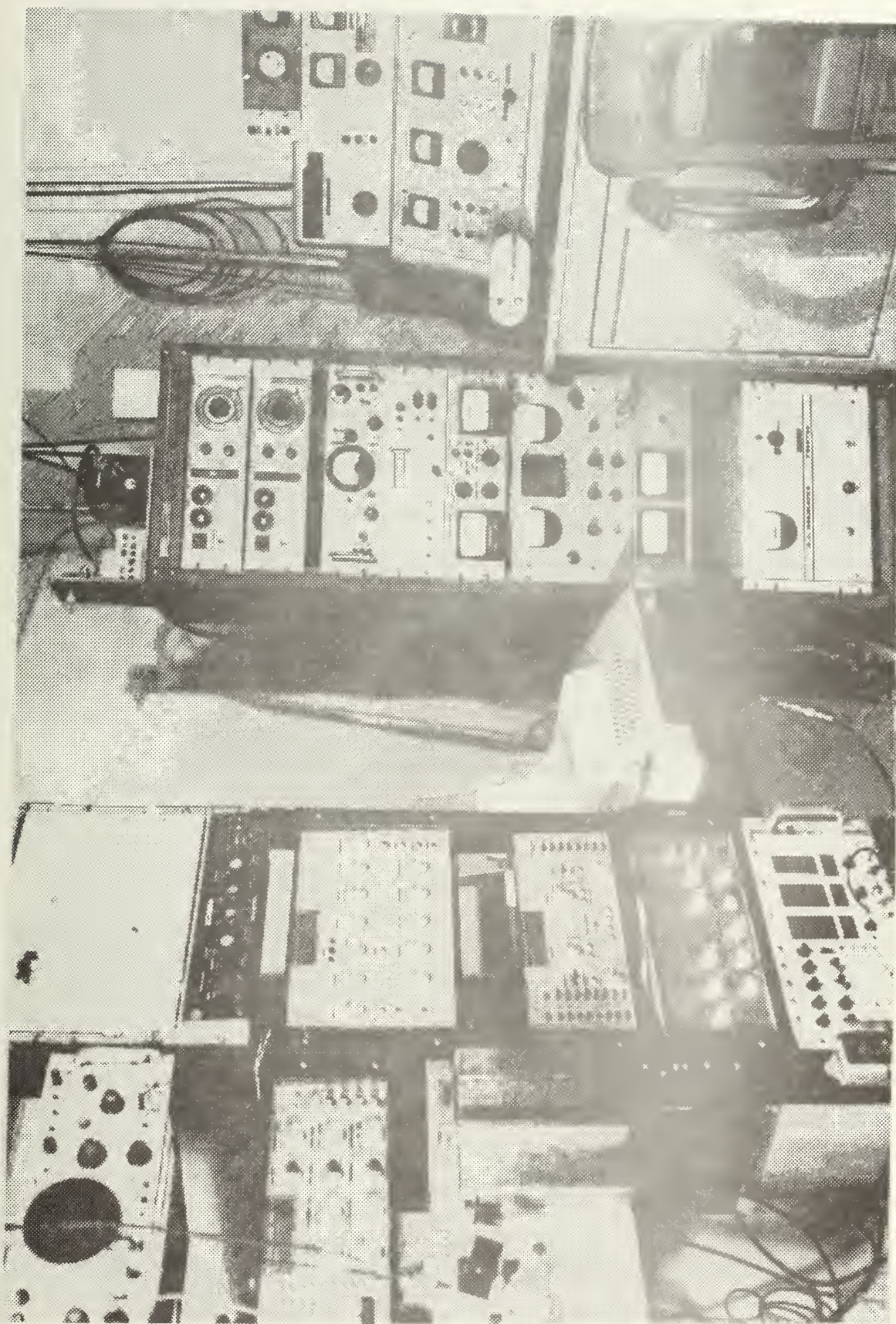


Figure 5. Control Console and Measuring Devices.

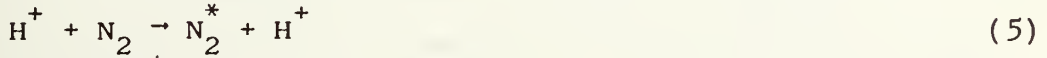
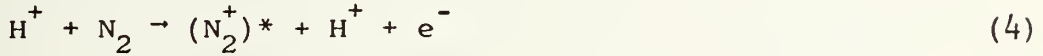




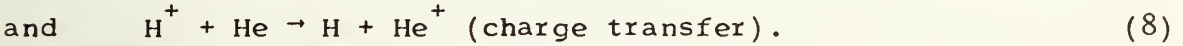
### III. THEORY

#### A. BACKGROUND

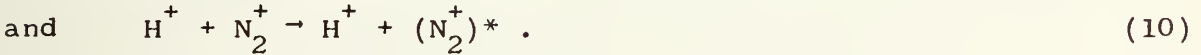
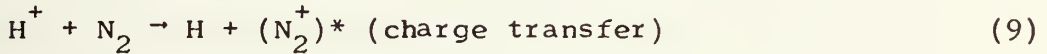
The primary nitrogen excitation reactions occurring in the collision chamber upon proton impact are:



The primary helium excitations expected to occur are:

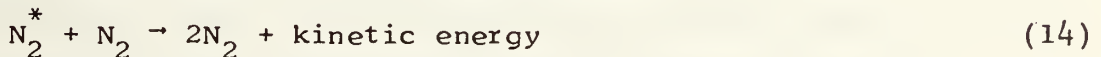
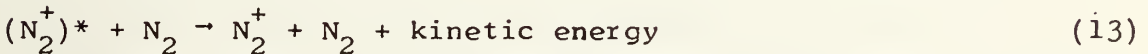
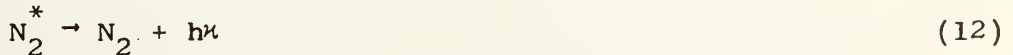
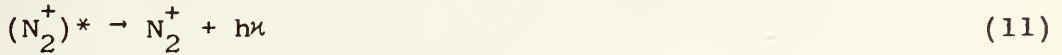


We can also consider the following excitation processes by proton impact:



The population of  $N_2^+$  is estimated to be too low for reaction (10) to be considered.

The primary de-excitation reactions of  $B^2\Sigma_u^+$  in  $N_2^+$  and  $C^2\Pi_u$  in  $N_2$  are:





The general equation [3] is given by:

$$\begin{aligned} \frac{dN_k^*}{dt} = 0 = & p v N \sigma_k + \sum_{\ell > k} \lambda_{\ell k} N_{\ell}^* - \sum_{i < k} \lambda_{ki} N_k^* \\ & - \sum_j N_k^* k_j n_j + \sum_m n_m^* k_m n_k \end{aligned} \quad (16)$$

where,

a)  $p v N \sigma_k$  is the direct excitation to state  $k$  by proton excitation given in terms of excitation cross section  $\sigma_k$ , proton beam density  $p$ , target density  $N$ , and proton velocity  $v$ .

The excitation cross section  $\sigma_k$  is energy dependent but the proton energy was held constant through the experiment, thus  $\sigma_k$  was taken as a constant. The proton beam density and velocity  $v$  were also constant for a given run, hence, the whole term was constant and was assumed to be proportional to the partial pressure ( $P_N$ ) in the target chamber [1]. Then

$$p v N \sigma_k = R_k \sim P_N$$

b)  $\sum_{\ell > k} \lambda_{\ell k} N_{\ell}^*$  accounts for transitions from all higher excited states  $\ell$  into state  $k$ , given in terms of transition probabilities  $\lambda_{\ell k}$  of decay from state  $\ell$  into state  $k$ , and  $N_{\ell}^*$  the number density of excited particles in state  $\ell$ .

The states above state  $k$  can be populated by direct interaction with protons. Thus higher states can then decay to state  $k$  and thus serve as an additional mechanism for excitation to state  $k$ . These excitation processes were also assumed to be



proportional to the partial pressure, and the experimental data indicated that this was a valid assumption.

c)  $\sum_{i, k>i} \lambda_{ki} N_k^*$  represents radiative loss by transitions from state  $k$  to a lower state  $i$ , where  $\lambda_{ki}$  are transition probabilities, and  $N_k^*$  is the number density of excited particles in state  $k$ .

In this term it was assumed that  $\sum_{i, k>i} \lambda_{ki} = \lambda$  a constant for the spectral line under study.

d)  $\sum_j N_k^* k_j n_j$  accounts for collisional de-excitation with particle  $n_j$ , where  $k_j$  is the collisional de-excitation rate,  $N_k^*$  is the density of excited particles and  $n_j$  is the density of particles of type  $j$ .

The assumptions made in this term were that  $k_j$  was a sum over only two gases ( $N_2$  and He), and  $n_j$ , the number density of particles of type  $j$ , was proportional to the partial pressure of these particles, which followed directly from the ideal gas law [5]. It was also assumed that the number density of the particles in excited state  $k$ ,  $N_k^*$  was proportional to the photon intensity (I).

e)  $\sum_m n_m^* k_m n_k$  represents excitation to state  $k$  by collision with atoms or molecules in excited state  $n_m^*$ , where  $n_m^*$  was the density of particles in state  $m$ ,  $k_m$  was the collisional excitation reaction rate, and  $n_k$  was the density of target particles.

When the chamber was filled with nitrogen alone, this term could be neglected as was explained by Walters [2]. In the case of nitrogen-helium mixtures in the reaction chamber, this term



could not be neglected because of the marked increase in intensity, where the excited helium transferred its energy to nitrogen, giving an enhancement of the nitrogen spectrum.

## B. NITROGEN AS THE TARGET GAS

Under these assumptions and equilibrium conditions, equation (16) becomes

$$\frac{dN_k^*}{dt} = 0 = R_k - \lambda N_k^* - k_j n_j N_k^* \quad (18)$$

but

$$R_k = a P_{N_2}, \quad N_k^* = b I \quad \text{and} \quad n_j = \frac{P_j}{kT} \quad (19)$$

$$a P_{N_2} = b I + k_j \frac{b}{kT} P_{N_2} I \quad (20)$$

$$P_{N_2} = \frac{\lambda b}{a} I + \frac{k_j b}{a kT} P_{N_2} I \quad (21)$$

$$P_{N_2} = A I + B P_{N_2} I \quad (22)$$

where,

$$A = \frac{\lambda b}{a}, \quad B = \frac{k_j b}{a kT}$$

$P_{N_2}$  represents the pressure of Nitrogen

Equation (22) can be modified to:

$$I = \frac{P_{N_2}}{A + B P_{N_2}} = \frac{P_{N_2}}{A(1 + \frac{B}{A} P_{N_2})} \quad (23)$$

The constant A is a constant of proportionality and depends only on the experimental set up. The constant B/A is a function of the decay constant and the de-excitation reaction rate  $k_j$ . The value of B/A was obtained from Smelley's work [1].

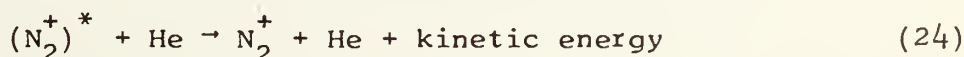




### C. NITROGEN WITH HELIUM PRESENT

The initial experimental results of nitrogen mixed with helium indicated that the presence of helium does not act as a quenching agent for  $N_2^+$  and  $N_2$  states as was observed in the experiment performed by Walters [2] and Smelley [1], but on the contrary gives an increase in the intensity of the lines in question, showing clearly that the collisional de-excitation rate is small compared to the collisional de-excitation rate in the case of nitrogen mixed with carbon dioxide or oxygen. Furthermore, as is shown in Figures 6 and 7 the intensity of the first negative band  $v = 0$  to  $v' = 0$  ( $\lambda = 3371.3$  A), and the second positive bands  $v = 0$  to  $v' = 1$  ( $\lambda = 4278.1$  A), and  $v = 0$  to  $v' = 0$  ( $\lambda = 3914.4$  A) increased as the partial pressure of helium was increased. Also it was observed that as the partial pressure of nitrogen decreased the intensity of nitrogen lines increased. This is a very surprising effect, especially for a partial pressure of nitrogen of 10 Torr as is shown in Figures 6 and 7. It is believed that the energy of the excited helium is transferred to nitrogen, which agrees with the results of the nitrogen-helium spectrum taken by Smelley, who noted the enhancement of the molecular nitrogen bands in the presence of helium. It also agrees with the results of Tullington [6] who made numerous unsuccessful attempts to obtain a pure helium spectrum. He observed primarily a nitrogen spectrum unless he had very pure helium.

The de-excitation reaction of  $(N_2^+)^*$  and  $N_2^*$  systems are as in equations (11), (12), (13), (14), and (15) as well as by:





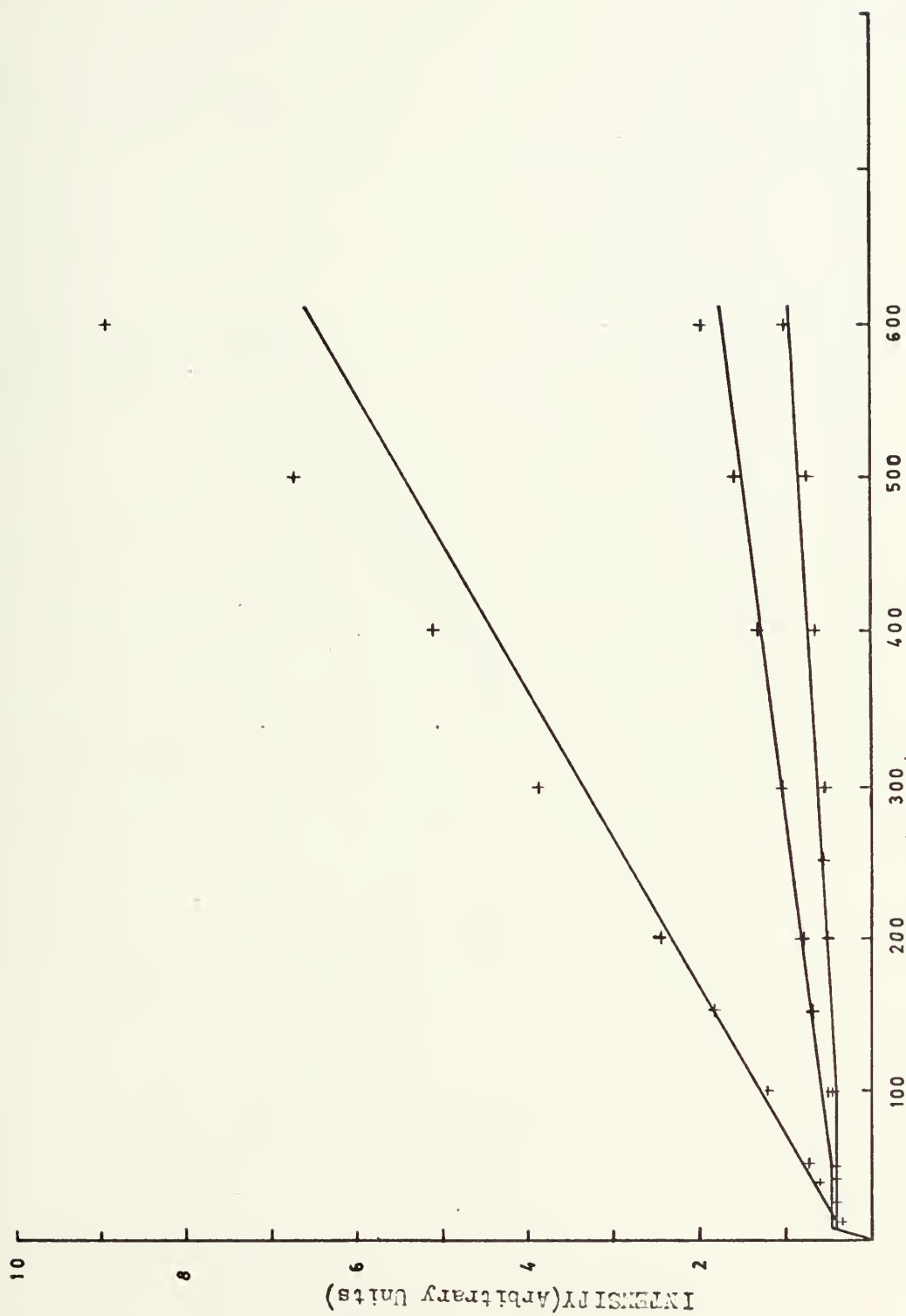


FIGURE 6. Intensity versus Pressure (3914.4 Å). H<sup>+</sup> on 10 torr N<sub>2</sub>, Balance He. H<sup>+</sup> on 50 torr N<sub>2</sub>, Balance He. H<sup>+</sup> on 100 torr N<sub>2</sub>, Balance He.



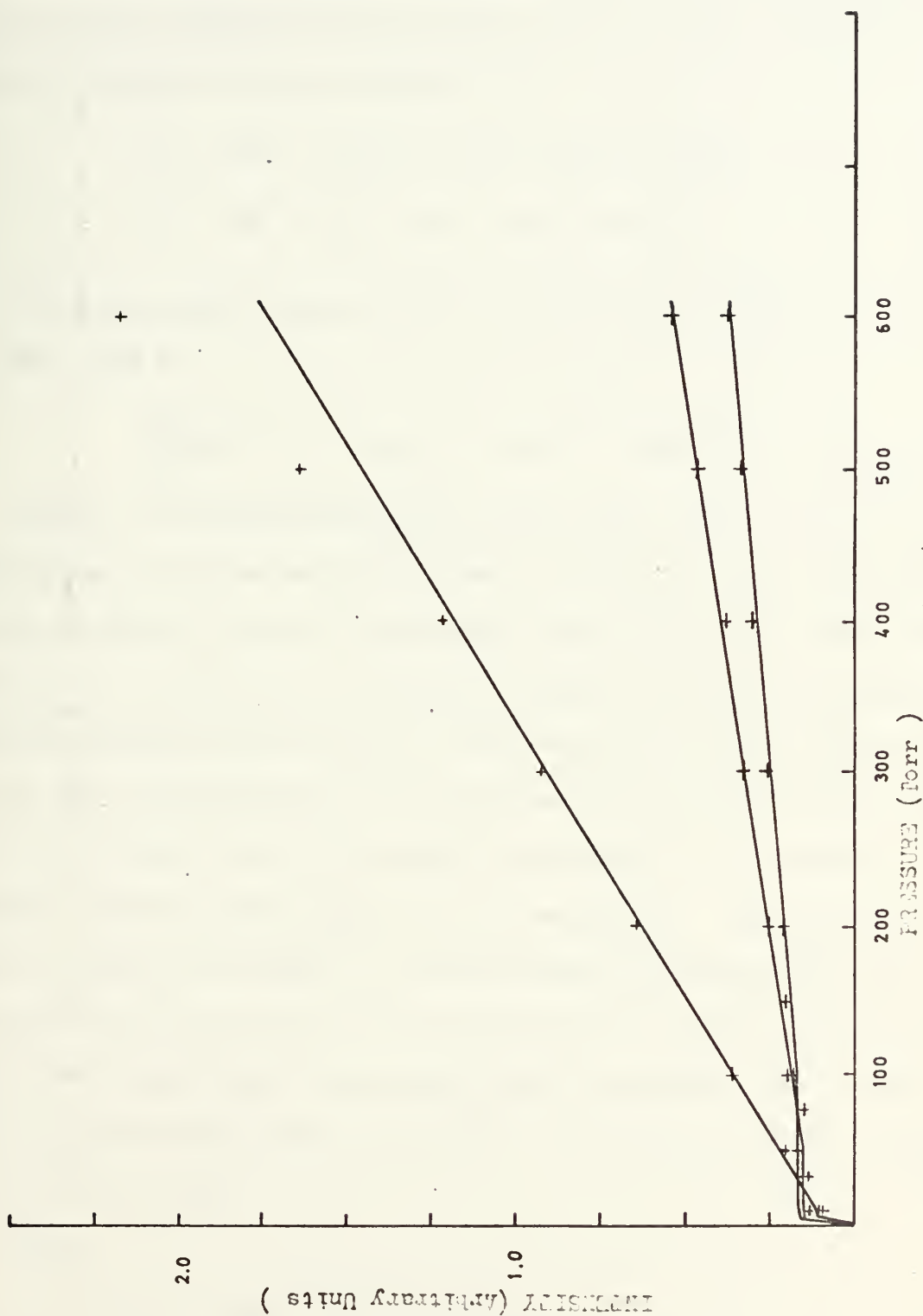
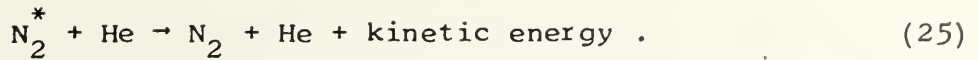
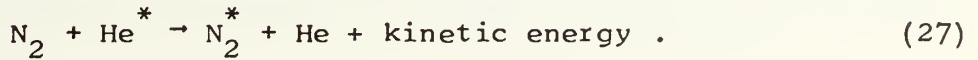
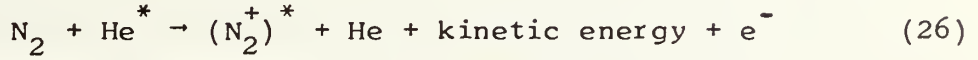


FIGURE 7. Intensity versus Pressure (4278.1 Å).  $H^+$  on 10 torr  $N_2$ , Balance He.  $H^+$  on 50 torr  $N_2$ , Balance He.





It is believed that these reactions proceed at a very low rate, and the main mechanisms which produce the increase in the intensity of the nitrogen lines are governed by:



From the general equation (16) the behavior of the helium can be described by

$$\frac{dn}{dt}(He^*) = 0 = R(He) - n(He)^* - n(He)^* n(N_2) k_{HeN_2} . \quad (28)$$

Where, as was explained before, the first term is the direct excitation of helium by proton impact to state t, the second term is the radiative loss by transitions from state t to a lower state expressed in terms of the transition probability and the number of excited helium atoms in state t, the third term accounts for the collisional de-excitation of the excited helium by nitrogen, and the fourth term is the collisional de-excitation of helium by helium. The last term appears to be negligible. Because no helium lines were evident in the spectrum of helium mixed with nitrogen [1,6], the third term of equation (16) can be neglected, and for the same reason the second term of equation (28), which are considered much smaller than the collisional de-excitation of helium by nitrogen.

Then,

$$R_{He} = n_{He}^* n_{N_2} k_{HeN_2} . \quad (29)$$





But,  $R_{\text{He}}$  is proportional to the helium pressure, then

$$C P_{\text{He}} = n_{\text{He}}^* n_{\text{N}_2} k_{\text{HeN}_2}. \quad (30)$$

Now in a mixture of nitrogen and helium, the collisional de-excitation of helium due to collision with nitrogen and the collisional excitation of nitrogen due to collision with excited helium are assumed to be equal or at least proportional, reactions (26) and (27).

Then equation (22) can be expressed

$$P_{\text{N}_2} = A I + B P_{\text{N}_2} I - C P_{\text{He}} \quad (31)$$

or

$$I = \frac{P_{\text{N}_2} + C P_{\text{He}}}{A(1 + \frac{B}{A} P_{\text{N}_2})}. \quad (32)$$



#### IV. RESULTS

In this study it has been shown that the intensity of nitrogen bands increased when nitrogen was mixed with helium indicating a transfer of the energy of excitation from the helium to nitrogen. Whereas, it was observed that the nitrogen band intensity decreased when nitrogen was mixed with carbon dioxide or oxygen showing clearly a collisional de-excitation phenomenon.

It was observed that for the second positive ( $C^3\Pi_u \rightarrow B^3\Pi_g$ ) and the first negative ( $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ ) transitions, as the partial pressure of nitrogen decreased the intensity of the nitrogen bands increased. Figure 6 and 7 show this effect for the first negative bands, transitions  $v = 0$  to  $v' = 0$  ( $\lambda = 4278.1 \text{ A}$ ).

Equation (23) was used to obtain intensity for the case of nitrogen alone. This equation agreed with the experimental data obtained in this work. The constant  $B/A$  was obtained from Smelley's work.

The assumptions made in arriving at equation (30) seem to be valid in light of the close correlation between the experimental results and the theoretical equations. In the case of helium mixed with nitrogen there are two excitation processes, direct excitation by proton excitation and collisional transfer of excitation from helium to nitrogen.

The theoretical curves of pressure versus intensity for  $\lambda = 3914.4 \text{ A}$ ,  $v = 0$  to  $v' = 0$  transition shown in Figure 6 and for  $\lambda = 4278.1 \text{ A}$ ,  $v = 0$  to  $v' = 1$  transition shown in Figure 7





were obtained using the value of  $B/A$  from Smelley's work, and the value  $C = 0.251 \pm 0.001$  obtained for the best fit to the experimental data. The slope of the curves increase for a decrease in the partial pressure of nitrogen. The curves of experimental data tend to deviate from the linear behavior at high partial pressures of helium, especially for a partial pressure of nitrogen of ten Torr.

A run was made where the collision chamber was filled with helium at 20 Torr pressure adding nitrogen gas in increments, using the same procedure as explained in the experimental procedure section. Figure 8 shows that the values of the constants  $B/A$  and  $C$  are in complete agreement with the experimental observations and in this way with the assumptions made to obtain the Equation (30). The de-excitation of the nitrogen bands as the partial pressure of nitrogen was increased was predicted.

The same phenomenon was observed for the second positive band, transition  $v = 0$  to  $v' = 0$  ( $\lambda = 3371.3 \text{ \AA}$ ) where the experimental results are shown in Figures 9, 10, and 11.



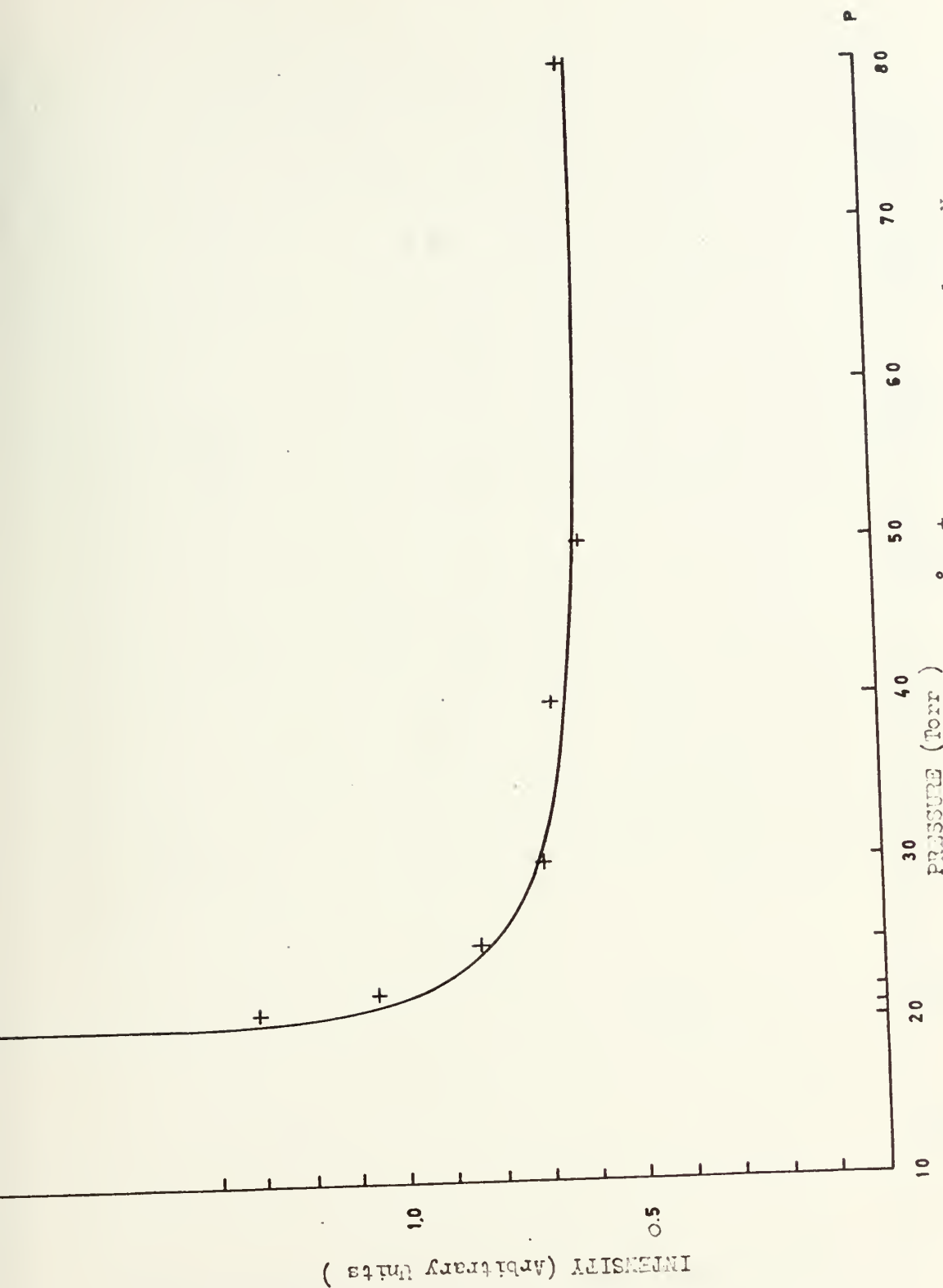


FIGURE 3 . Intensity versus Pressure. (4278.1 Å).  $\text{H}^+$  on 20 torr He, Balance  $\text{N}_2$ .



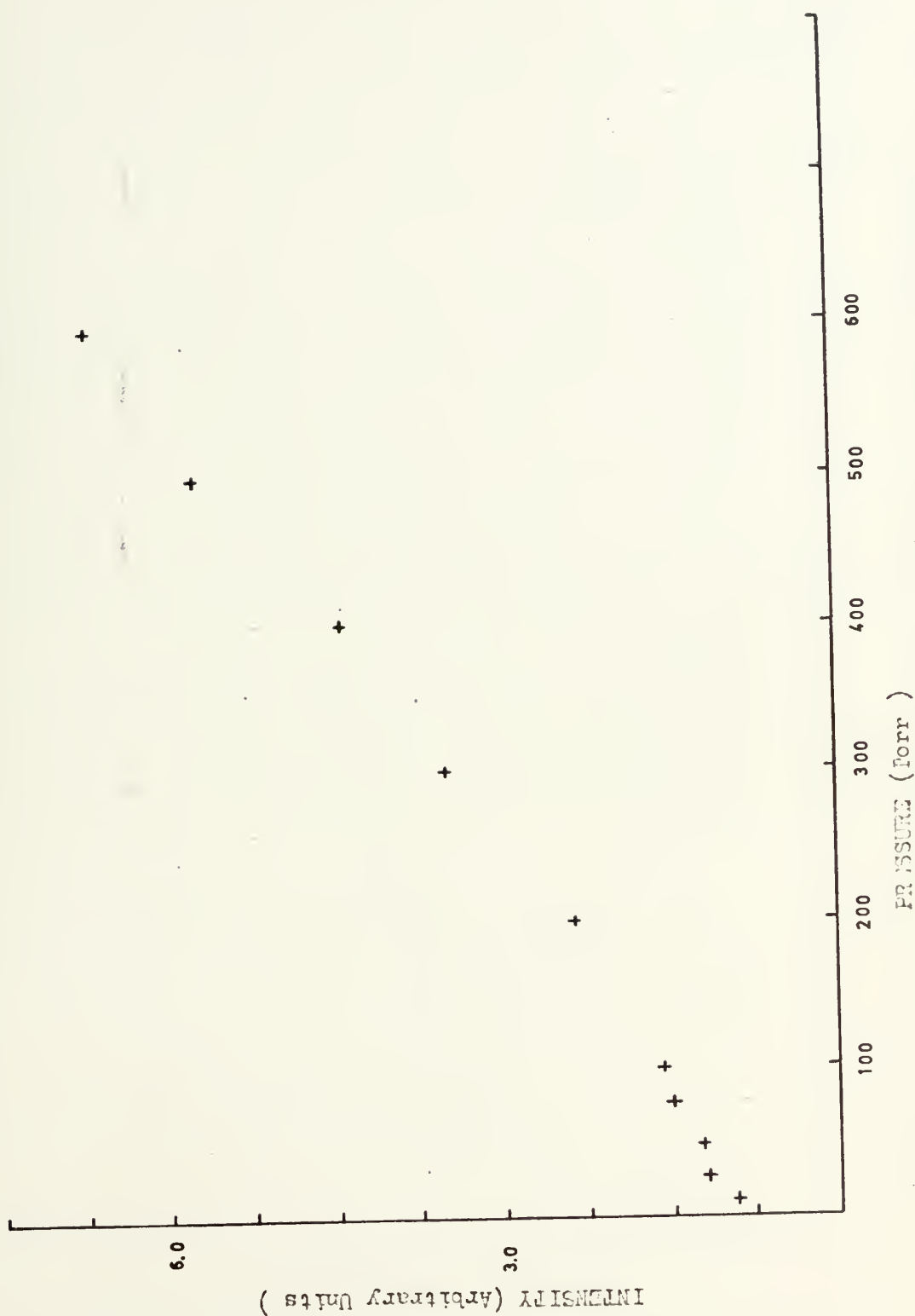


FIGURE 9 .Intensity versus Pressure (3371.3 Å).H<sup>+</sup> on 10 torr N<sub>2</sub>, Balance He.





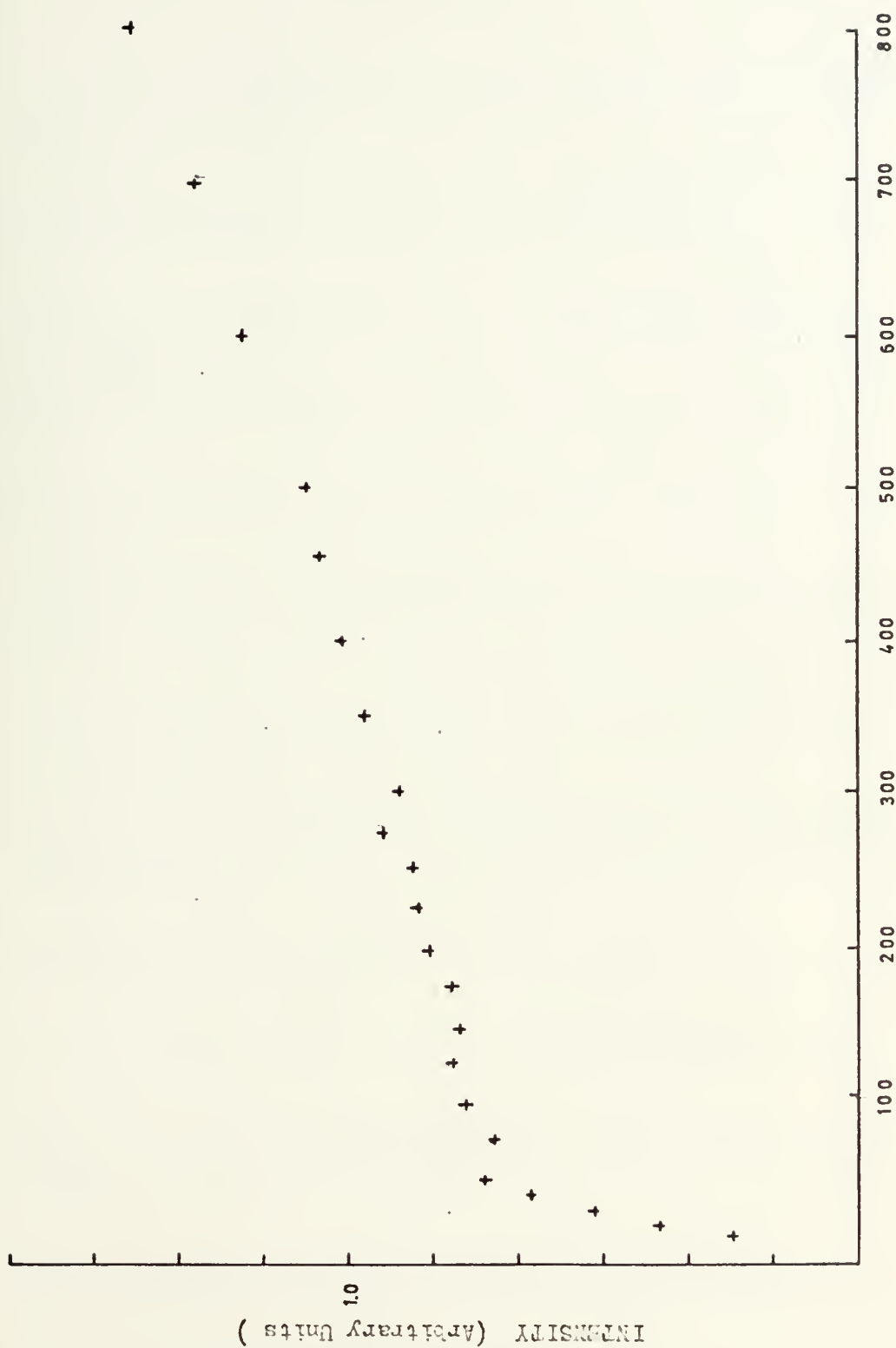


FIGURE 10. Intensity versus Pressure ( $3371.3 \text{ \AA}$ ).  $\text{H}^+$  on 50 torr  $\text{N}_2$ , Balance He.



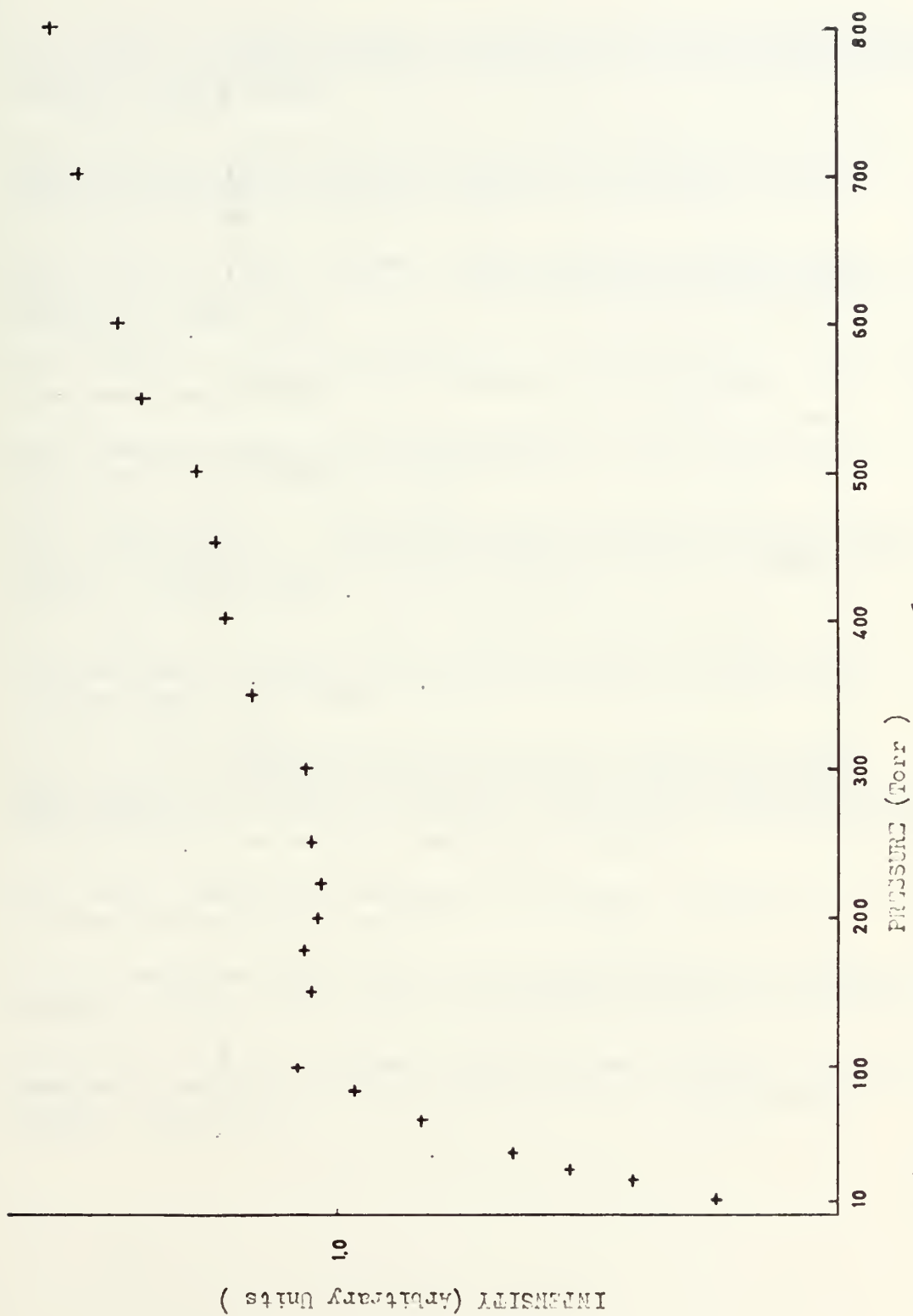


FIGURE 11. Intensity versus Pressure (3371.3 Å).  $H^+$  on 100 torr  $H_2$ , Balance He



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13. ABSTRACT

Nitrogen and nitrogen-helium mixtures were excited by 1.4 MeV protons from a Van de Graaff generator. Intensity versus pressure data from 1 to 600 Torr were recorded and plotted for the first negative band, transitions  $v = 0$  to  $v' = 0$  ( $\lambda = 3914.4$  A), and  $v = 0$  to  $v' = 1$  ( $\lambda = 4278.1$  A) and second positive transition  $v = 0$  to  $v' = 0$  ( $\lambda = 3371.3$  A) of molecular nitrogen. The theoretical equation from Smelley's work [1] for nitrogen alone was verified and the coefficient  $B/A$  from Smelley's work was used in this work. A theoretical equation was derived for intensity as a function of partial pressure of nitrogen and helium, which was shown to agree quite well with the experimental data and the coefficient of enhancement  $C = 0.251 \pm 0.001$  was obtained. As the experimental data shows, a very interesting collisional excitation process was observed, which was not observed for nitrogen-oxygen and nitrogen-carbon dioxide mixtures.



- Hydrogen
- Hydrogen-Helium
- Cross-section
- Collisional excitation
- Excitation rate









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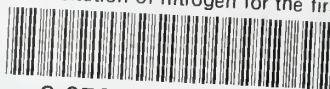
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